IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants:	Donald R. Huffman, et al.	**Examiner:**	Tsang Foster, S.N.
Serial No.:	08/236,933	**Art Unit:**	1745
Filed:	May 2, 1994	**Docket:**	7913ZAZY
For:	NEW FORM OF CARBON		

Confirmation No.: 4115

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION OF ADAM DARWISH PURSUANT TO 37 C.F.R. §1.132

Sir:

I, ADAM DARWISH, declare and say as follows:

1. I am currently a tutor of organic chemistry in the Chemistry Department at Sussex University, England. I was awarded a Ph.D. in Physical Organic Chemistry in 1992 and have continued working at Sussex University as a Research fellow until 1999, when I received a promotion to senior research fellow. I have conducted research in the area of fullerenes for several years. This research includes, but is not limited to, the preparation and purification of gram quantities of fullerenes, developing new fullerene-producing reactions, and improved chromatographic separation of C_{60} and C_{70}. I am appending hereto an abbreviated version of my *curriculum vitae*, which summarizes my experience in the fullerene area.

2. In preparing this Declaration, I have read and reviewed the contents of USSN 08/236,933 in its entirety (" '933 application"), especially the description therein of the

preparation of fullerenes, including C_{60} and C_{70}, and especially the procedure for separating the fullerenes from the soot described therein, especially in Example 1 thereof.

3. I was introduced to applicants' attorney by Dr. Harold Kroto.

4. I was requested by applicants' attorney to separate fullerenes, including C_{60} and C_{70}, from the soot sample prepared by Dr. Terrones, which was forwarded to me.

5. The procedures described herein were either conducted by me or under my direct supervision and control.

6. The procedure used for separating the fullerenes from the soot utilized common separation techniques that were described in the '933 application or known and routine to one of ordinary skill in the art on August 30, 1990.

7. The same procedure was utilized for separating the fullerene from the soot produced at the lower pressure of 100 torr and at the higher pressure at 2 atm. More specifically, the samples of soot were extracted using a soxhlet extractor utilizing toluene as the solvent. Approximately, 10% of the soot sample was collected as soot extract. In the sample containing 1 gram of soot, i.e., the soot that was prepared from the vaporization of graphite at 100 torr using a current of about 100 amps, the soot extract was separated into the various fullerene fractions using preparative HPLC under the following conditions: Cosmosil 5μm PYE column (250mm x 10mm), HPLC-grade toluene as the solvent, eluted from the column at a rate of 4 ml/min. and the UV detector was set at 285 nm wavelength. Those fractions having an absorbance at 285 nm were collected. See Exhibit 2.

8. The fullerene fractions were then purified by recycling using the conditions described in Paragraph 7 herein.

9. A fullerene fraction, consisting of 65 mg of pure C_{60} crystals, another fullerene fraction consisting of 15 mg of C_{70} crystals and a third fullerene fraction of 7 mg of higher fullerenes comprised of C_{76} (1.2 mg), C_{78} (1.8 mg, two isomers) C_{84} (2.5 mg), C_{86} (0.5 mg), and C_{90} (1.0 mg, two isomers) together with $C_{70}O$ (2 mg), were collected from the 1 gram of soot prepared from the vaporization of graphite at 100 torr using a current of about 100 amp. Each of the fractions contained enough material to be seen with the naked eye.

10. The identity of each of the fullerenes in each of the fractions was verified by the mass spectra.

11. I have attached hereto copies of the mass spectra of each of the fullerenes isolated from the vaporization of graphite at 100 torr using a current of about 100 amps and the HPLC tracings of each isolated fullerene. In addition, I have attached the photographs of samples of each of the separated fullerenes dissolved in toluene and photographs of the crystals of each of these fullerenes obtained from evaporation of exactly the half volume of the toluene solution obtained except for C_{86} where all the toluene solution was evaporated to dryness. (see Exhibits 3-12).

12. Exhibit 3 includes the mass spectrum of a sample of C_{60} (3)(a) and the HPLC tracing of the C_{60} fraction (3)(b). Exhibit 3 further includes a photograph of a sample of C_{60} in solution in toluene (3)(c) and a photograph of the C_{60} crystals obtained from the evaporation of the toluene (3)(d). The mass spectrum confirmed the identity of the C_{60}, and both the mass spectrum and the HPLC tracing showed that the product is quite pure. As indicated hereinabove, 65 mg of the C_{60} crystals were recovered, which amount can be seen with the naked eye. Moreover, as further shown from the photograph in (3)(d), C_{60} is present in sufficient amounts to be seen with the naked eye.

13. Exhibit 4 depicts the mass spectrum of the C_{70} fraction (4)(a) and the HPLC tracing of the C_{70} fraction (4)(b). The mass spectrum confirms the identity of C_{70}, while the mass spectrum and the HPLC tracing show that the sample is quite pure (Exhibit 4). Exhibit 4 further includes a photograph of a sample of the C_{70} dissolved in toluene (4)(c) and a photograph of C_{70} after evaporation of the toluene (4)(d). As indicated hereinabove, 15 mg of C_{70} were collected, which also can be seen with the naked eye. As further shown by the photograph in Exhibit (4)(d), the C_{70} crystals were present in sufficient amounts to be seen with the naked eye.

14. The remaining fullerenes ("higher fullerenes") were present in a total amount of 9 mg, and the higher fullerenes in total were present in amounts sufficient to be seen with the naked eye.

15. Exhibit 5 shows the mass spectrum of $C_{70}O$, the HPLC tracing of the product and a photograph of $C_{70}O$ solution in toluene. Again, the mass spectrum confirms the identity of the product, while the mass spectrum and HPLC tracing show that it is quite pure.

16. Exhibit 6 depicts the mass spectrum of C_{76}, the HPLC tracing of C_{76}, and a photograph of C_{76} in solution in toluene. From the mass spectrum and the HPLC tracing, the product is quite pure. The mass spectrum confirms the identity of the product.

17. Exhibit 7 depicts the mass spectrum of C_{78}, the HPLC tracing of the two isomers of C_{78} that were obtained from the soot, and a photograph of C_{78} in solution in toluene. The mass spectrum confirms that the product is C_{78}, and the HPLC tracing clearly shows that 2 isomers were formed.

18. Exhibit 8 depicts the mass spectrum of C_{84}, the HPLC tracing of C_{84} and a photograph of C_{84} dissolved in toluene. The mass spectrum confirms the identity of the product as C_{84} and the mass spectrum and the HPLC tracing show that the product is quite pure.

19. Exhibit 9 depicts the mass spectrum of C_{86}, the HPLC tracing of same, and a photograph of C_{86} dissolved in toluene. The identity of the product is confirmed by the mass spectrum, and the mass spectrum and the HPLC tracing show that the product is quite pure.

20. Exhibit 10 depicts the mass spectrum of C_{90}, the HPLC tracing of two isomers of C_{90} and a photograph of C_{90} dissolved in toluene. The mass spectrum confirmed the identify of the product and the HPLC tracing shows that two isomers of C_{90} were collected.

21. Exhibit 11 is a photograph of various fullerenes, i.e, C_{60}, C_{70}, C_{76}, C_{78}, C_{84}, C_{86}, and C_{90} (that were obtained from the soot produced from the vaporization of graphite at 100 torr) dissolved in toluene.

22. Exhibit 12 consists of photographs of higher fullerene products, which were isolated from the soot produced from the vaporization of graphite at 100 torr, in solution in toluene and the solids of same obtained from the evaporation of exactly the half volume of the toluene solution obtained except for C_{86} where all the toluene solution was evaporated to dryness. The bottom portion of Exhibit 12 depicts each of these higher fullerene products. As can be seen by the photographs, the C_{76}, C_{78}, C_{84}, C_{86} and C_{90} produced from the soot obtained from the vaporization of graphite at 100 torr can be seen with the naked eye.

23. I also separated the second sample comprised of 100 mg. of soot that was produced at the higher pressure of 2 atm using the same technique as described in Paragraphs 7 and 8. Toluene (100 ml) was used as the solvent for extraction using a soxhlet extractor, and 9 mg or 9% yield was obtained. The various products obtained from this soot are described in Paragraphs 24-26.

24. Attached as Exhibit 13 is the mass spectrum of C_{60} and photographs of a sample of C_{60} in solution in toluene obtained from the soot produced from the vaporization of graphite at

2 atm and the C_{60} crystals obtained from the evaporation of the toluene. The mass spectrum confirmed the identity of C_{60}. As shown by the photograph in Exhibit 13, the C_{60} isolated (5.0 mg) from the 100 mg. of soot received from Dr. Terrones can be seen with the naked eye.

25. Attached as Exhibit 14 is the mass spectrum of C_{70}, and the photographs of a sample of C_{70} in solution in toluene obtained from the soot produced from the vaporization of graphite at 2 atm and C_{70} crystals (1.5 mg) obtained from the evaporation of the toluene. Again, as shown by the photograph, the C_{70} isolated from the 100 mg. of soot, which was sent by Dr. Terrones, can be seen with the naked eye.

26. Exhibit 15 depicts the mass spectrum of the higher fullerenes obtained when the vaporization of graphite was performed at 2 atm. The mass spectrum depicts that higher fullerenes up to C_{104} were detected by the mass spectrum. Exhibit 15 also depicts the photographs of a sample of higher fullerenes in solution in toluene and photographs of the crystals of the higher fullerenes (1.0 mg) obtained after evaporation of toluene. Thus, the higher fullerenes in total were present in the soot obtained from the vaporization of graphite at 2 atm in amounts sufficient to be seen with the naked eye.

27. Exhibit 16 consists of photographs of separate samples of C_{60}, C_{70} and the higher fullerenes, isolated from the soot prepared from the vaporization of graphite at 2 atm and 100 amps, dissolved in toluene and photographs of the crystals of C_{60}, C_{70} and the higher fullerenes obtained from evaporation of the toluene therefrom. Thus, as shown by the photographs, these crystals of C_{60}, C_{70} and higher fullerenes can be seen with the naked eye.

28. I further declare that all statements made herein of my own knowledge are true and that alls statements made on information and belief are believed to be true and further that these statements were made with the knowledge that willful false statements and the like so

made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: August 28, 2007

A. Darwish

Adam Darwish

Curriculum Vitae

Name: Adam Darwish

Date of Birth: 7th March 1958

Gender: Male

Nationality: British

Marital Status: Married

Address: 15 Sunnydale Avenue, Patcham, Brighton, East Sussex, BN1 8NR
Tel No.: (Home) 01273 501522
(Mobile) 07810768880
E-mail: kapg2@sussex.ac.uk

EDUCATION

D.Phil Physical Organic Chemistry (Sussex University, 1989-1992, UK)

BSc. Chemistry (Yarmouk University, 1977-1980, Jordan)

RESEARCH EXPERIENCE

2006-Present Tutor of Organic Chemistry, Chemistry Department, Sussex University.
1999-2006 Senior Research Fellow, Chemistry Department, Sussex University.
1992-1999 Research Fellow, Chemistry Department, Sussex University

Research involved the preparation and purification of gram quantities of fullerenes, and development of a new fullerene-producing reactor together with improved chromatographic separation of C_{60} and C_{70}. Carrying out a number of studies into new chemical reactions of C_{60}, C_{70}, C_{76}, C_{78} and C_{84} and fluorinated fullerenes ($C_{60}F_{18}$, $C_{60}F_{18}O_x$ and $C_{60}F_{20}$). These included reduction, oxidation, fluorinations, additions, electrophilic and nucleophilic addition/substitutions, which have resulted in a large number of publications **(71 publications)** in a variety of scientific journals.

1989-1992 D.Phil research student (gas phase elimination of some nitrogen-containing heteroaromatics)

SELECTED PUBLICATIONS

1. Fullerenes, **Adam D. Darwish**, *Annu. Rep. Prog. Chem., Sect. A: Inorg. Chem.*, 2007, **103**, 370 – 391.

2. Pyrolytic Trifluoromethylation of [76]-, [78]-, [84]-, and Aza[60]Fullerene with silver Trifluoroacetate; Evidence for Coordination of Fullerenes to Silver, **Adam D. Darwish**, A. K. Abdul-Sada, and R. Taylor, *Fullerene, Nanotubes, and Carbon Nanostructures,* 2006, **14**(1), 111.

3. Isolation of Two Seven-membered Ring C58 Fullerene Derivatives: $C_{58}F_{17}CF_3$ and $C_{58}F_{18}$. P. A. Troshin, A. G. Avent, **A. D. Darwish**, N. Martsinovich, A. K. Abdul-Sada, J. M. Street and R. Taylor, *Science*, 2005, **309**, 278.

4. Electrophilic Substitution by the Fluorofullerene $C_{60}F_{18}$, **A. D. Darwish**, A. G. Avent, A. K. Abdul-Sada, I. V. Gol'dt, I. Kuvytchko, P. B. Hitchcock and R. Taylor, *Chem Eur. J.*, 2004, **10**, 4523.

5. Electrophilic Substitution of $C_{60}F_{18}$ into Phenols: HF Elimination Between OH and a 1,3-Shifted Fluorine Giving Benzofurano[2',3':10,26]hexadecafluoro[60]fullerene and Derivatives, **A. D. Darwish**, A. G. Avent, J. M. Street and R. Taylor, *Org. Biomol. Chem.*, 2003, **1**, 1764.

6. $C_{60}F_{20}$: "Saturnene", an Extraordinary Squashed Fullerene. O. V. Boltalina, V. Yu. Markov, P. A. Troshin, **A. D. Darwish**, J. M. Street and R. Taylor, *Angew. Chem. Int. Ed.*, 2001, **40**, 787.

7. Novel Base-Catalysed Formation of Benzo[b]furano[60]- and –[70]Fullerenes. **A.D. Darwish**, A.G. Avent, H.W. Kroto, R. Taylor and D.R.M. Walton, *J. Chem Soc., Perkin Trans. 2,* 1999, 1983.

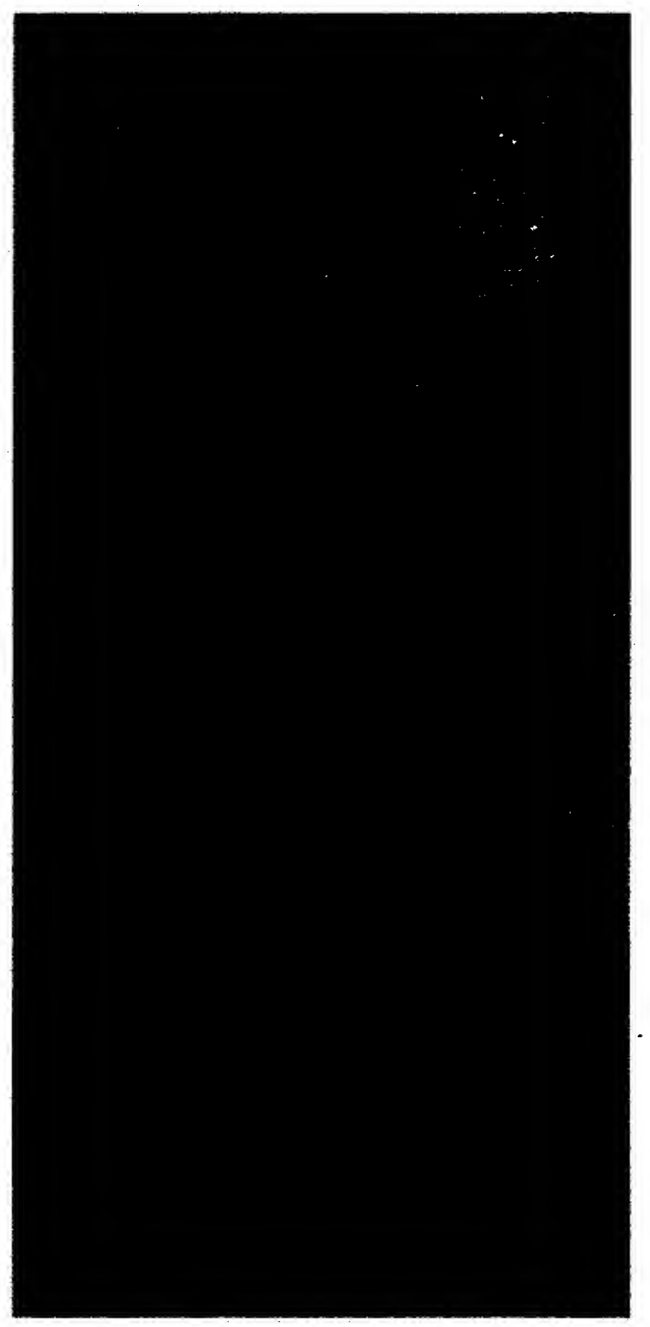
8. Hydrogenation of [76]-, [78]- and [84]Fullerenes. **A.D. Darwish**, H.W. Kroto, R. Taylor and D.R.M. Walton, *J. Chem Soc., Perkin Trans. 2*, 1996, 1415.

9. Preparation and ^{13}C NMR Spectroscopic Characterization of $C_{60}Cl_6$. P.R. Birkett, A.G. Avent, **A.D. Darwish**, H.W. Kroto, R. Taylor and D.R.M. Walton, *J. Chem Soc., Chem. Commun.*, 1993, 1230.

DARWISH EXHIBIT 2

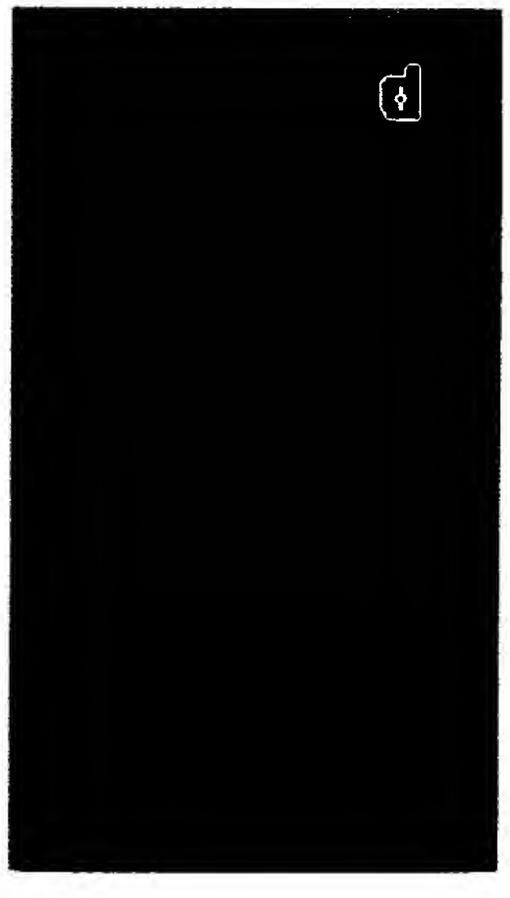
a

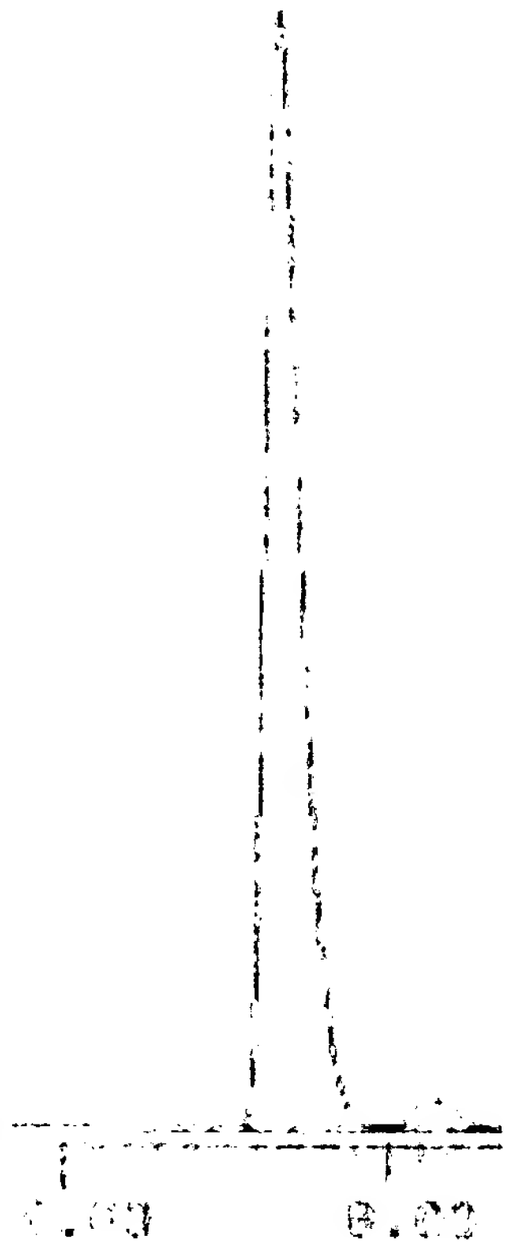
b



c

a. Soxhlet extractor
b. Soot extract in 100ml of toluene
c. Concentrated soot extract in 30ml of toluene





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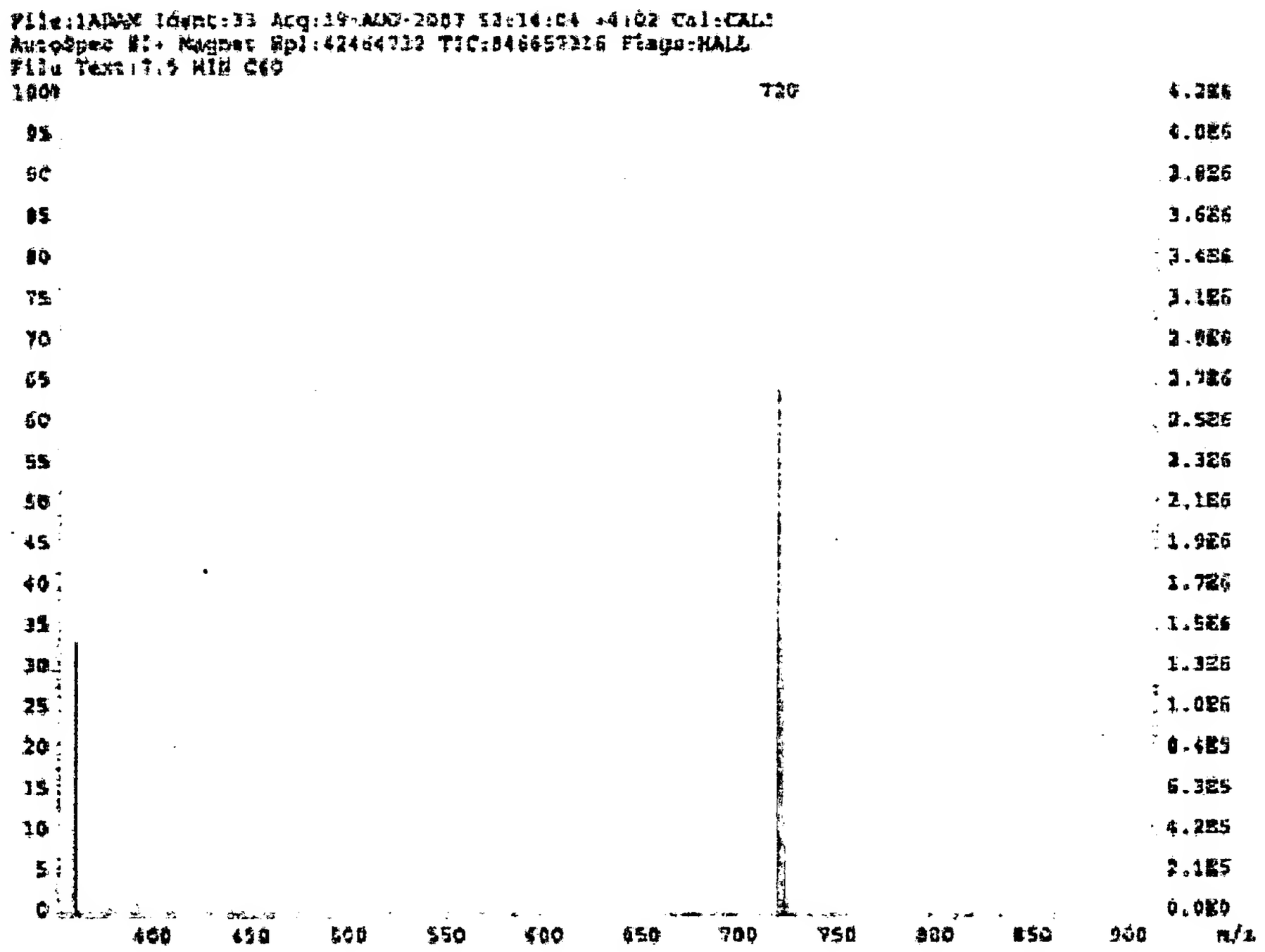
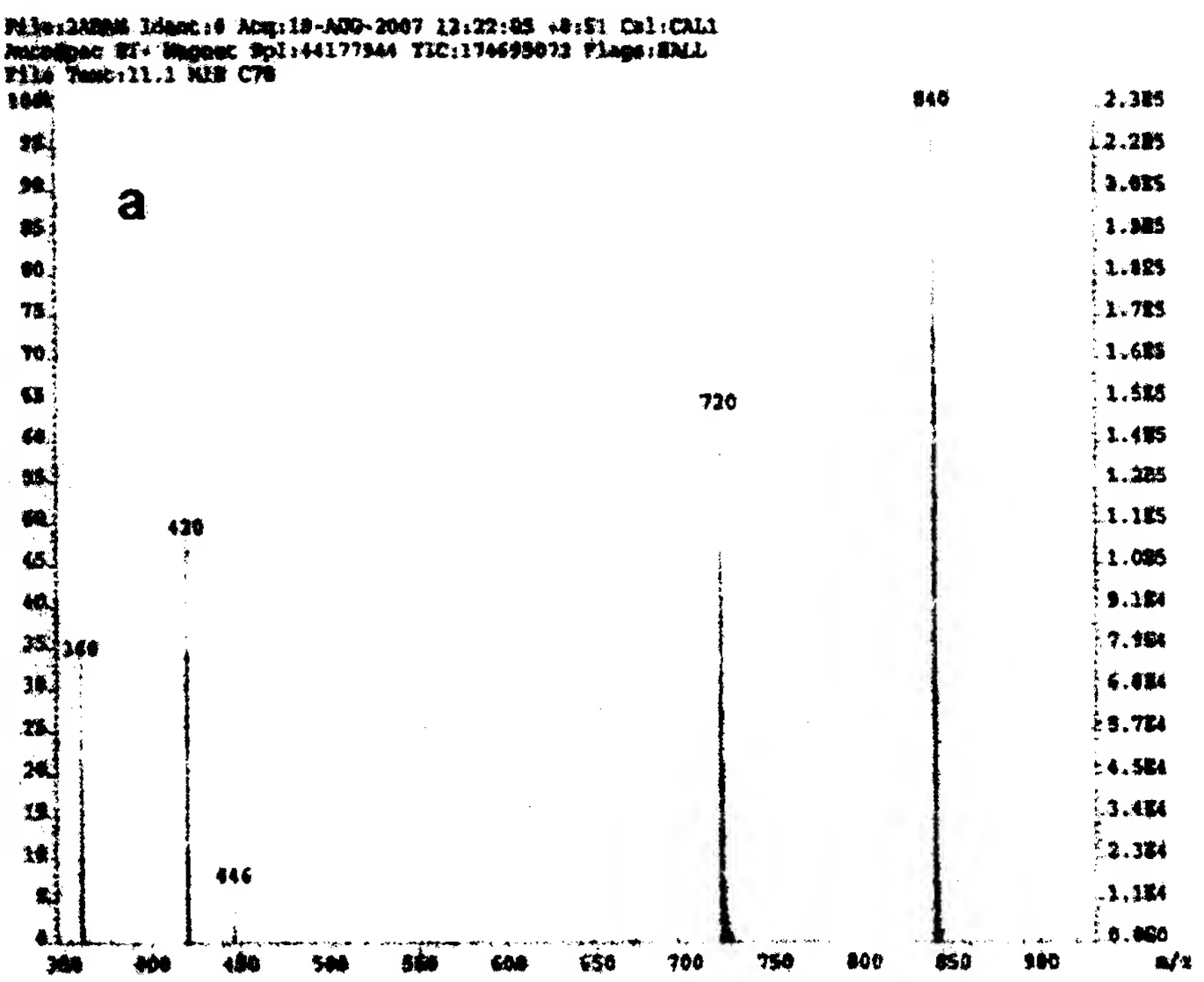


a

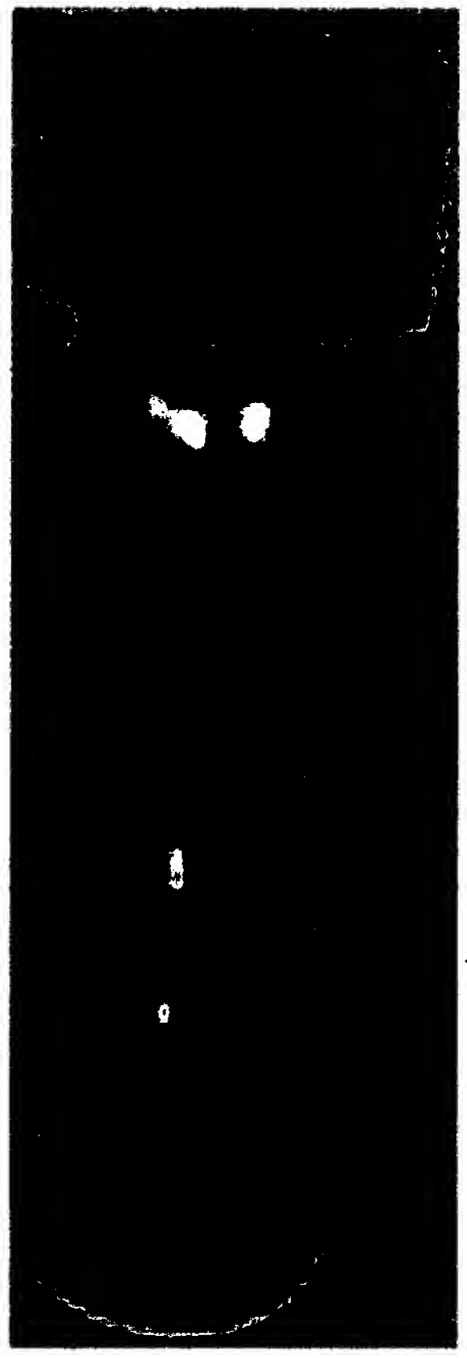
a. Mass spectra of C_{60}
b. HPLC traces (7.5 minute)
c. C_{60} solution in toluene
d. C_{60} crystals

c



a. Mass spectra of C_{70}
b. HPLC traces (11.1 minutes)
c. C_{70} solution in toluene
d. C_{70} crystals

DARWISH EXHIBIT 4



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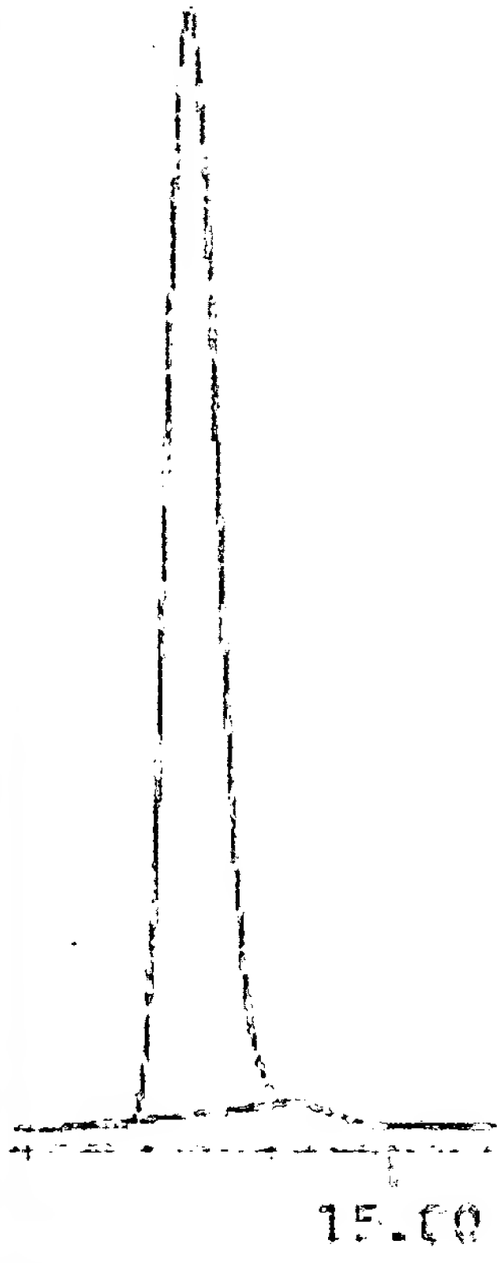

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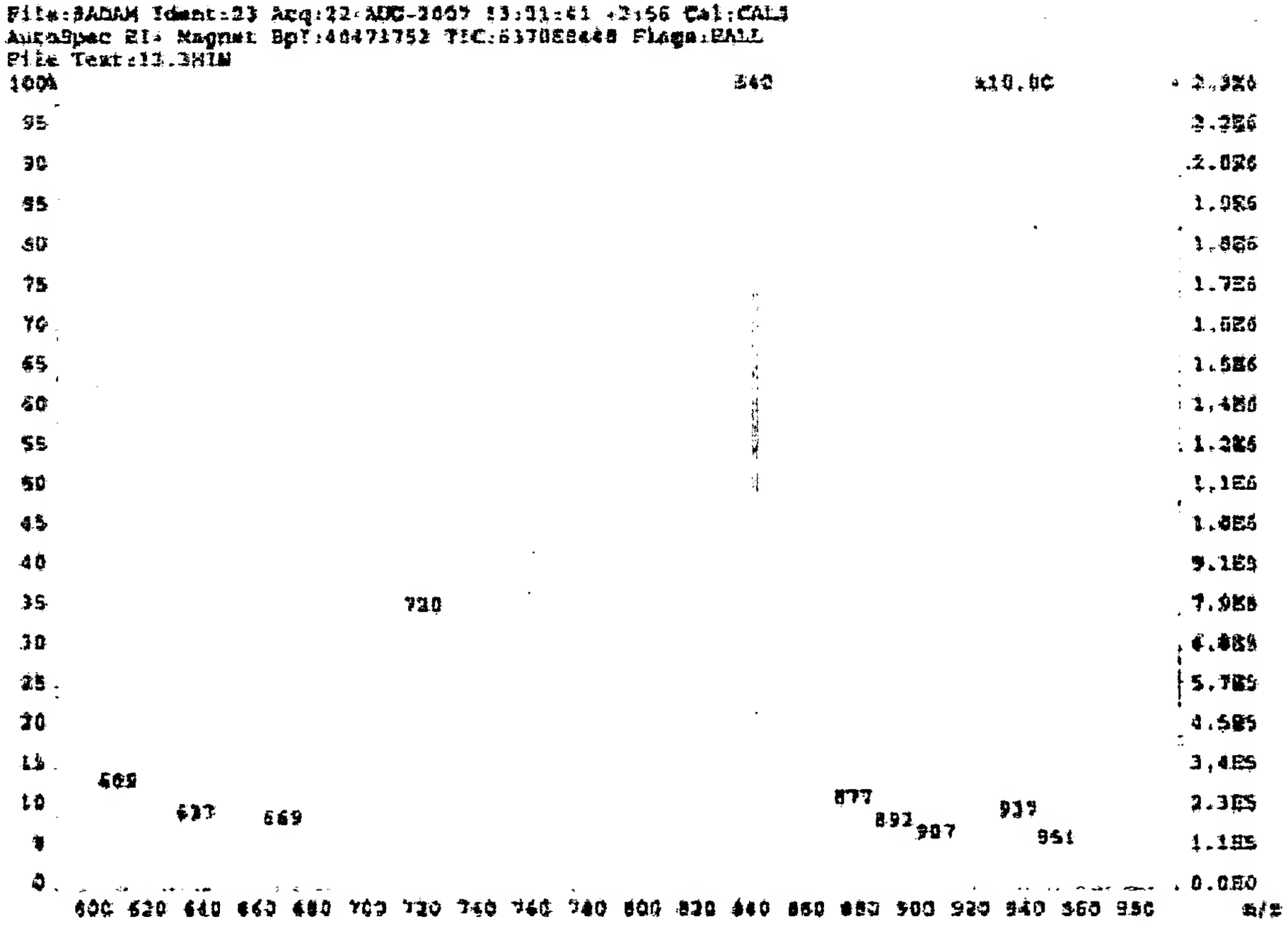
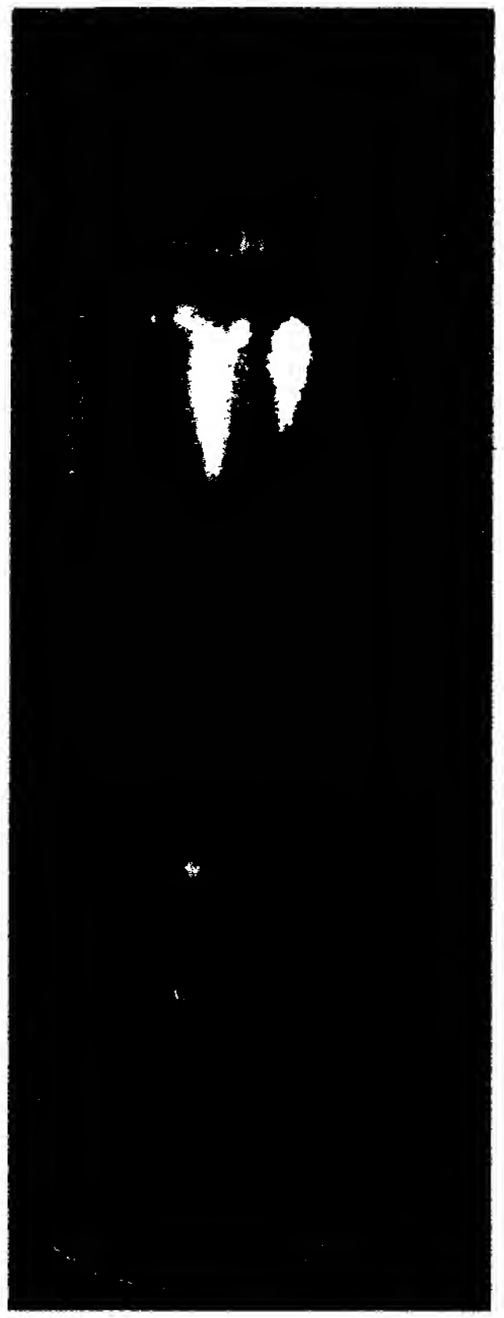


a

a. Mass spectra of $C_{70}O$.
b. HPLC traces (13.3 minutes.)
c. $C_{70}O$ solution in toluene



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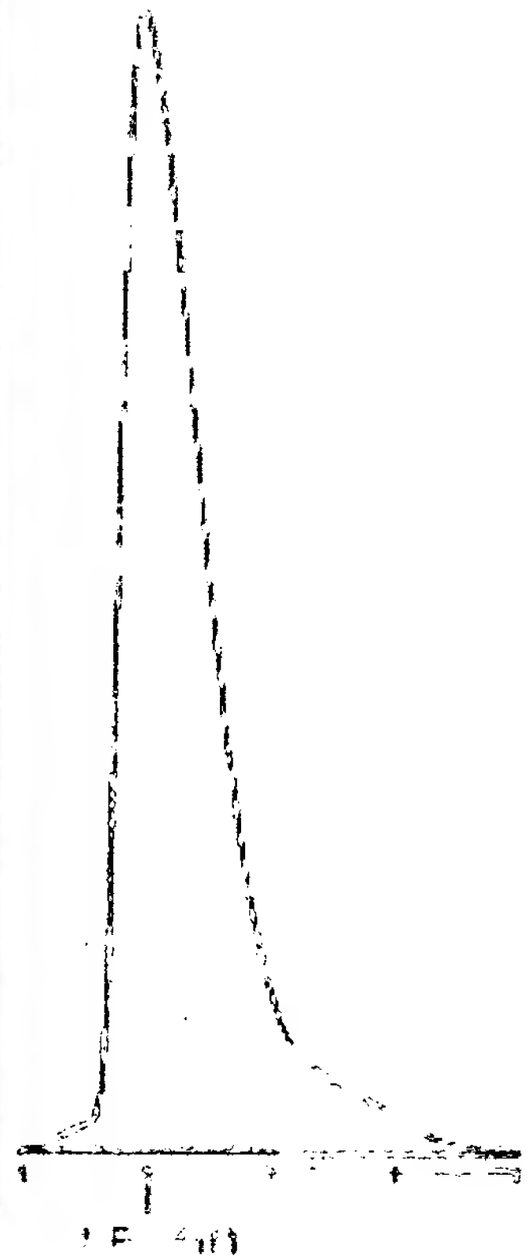

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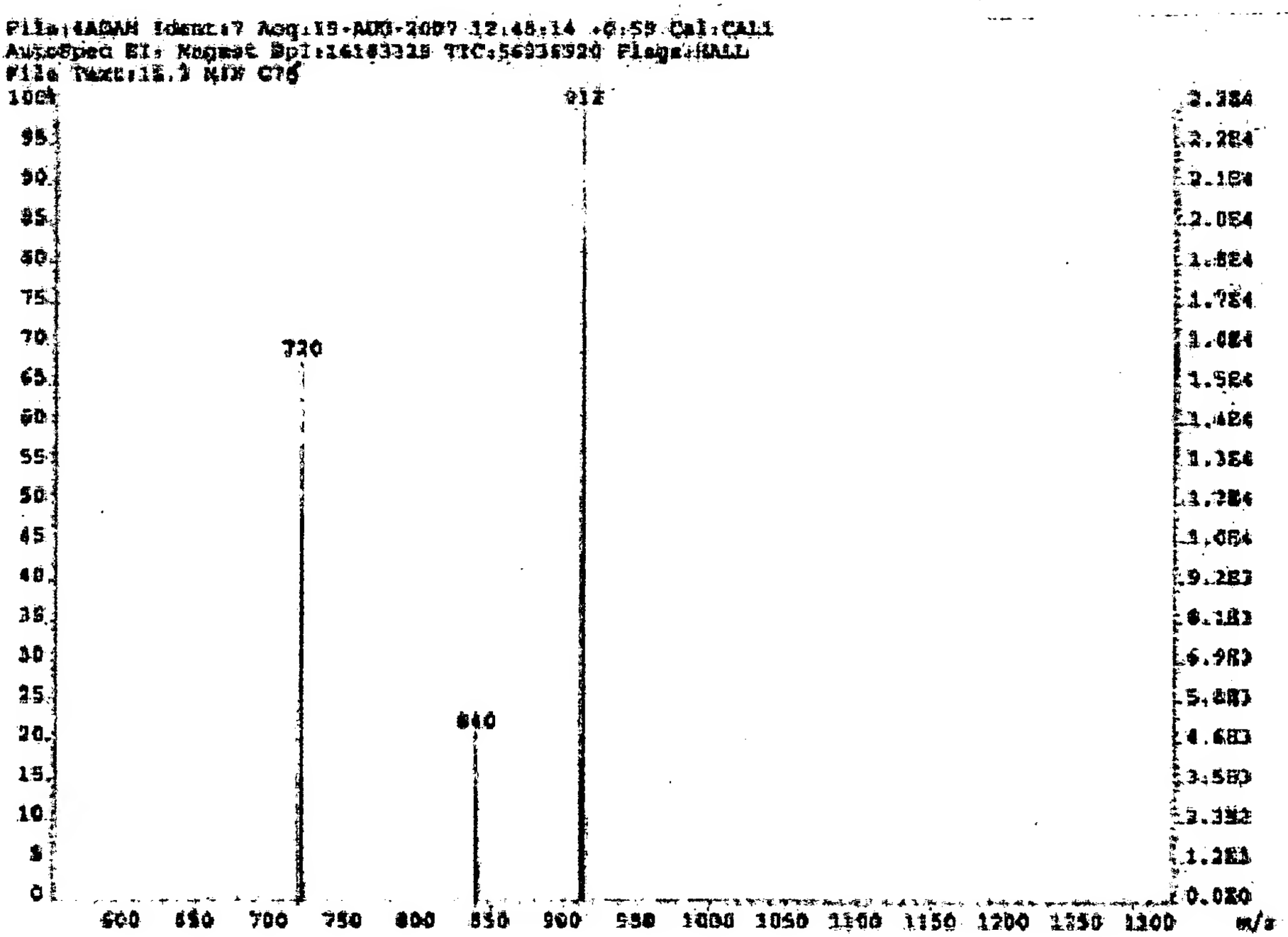
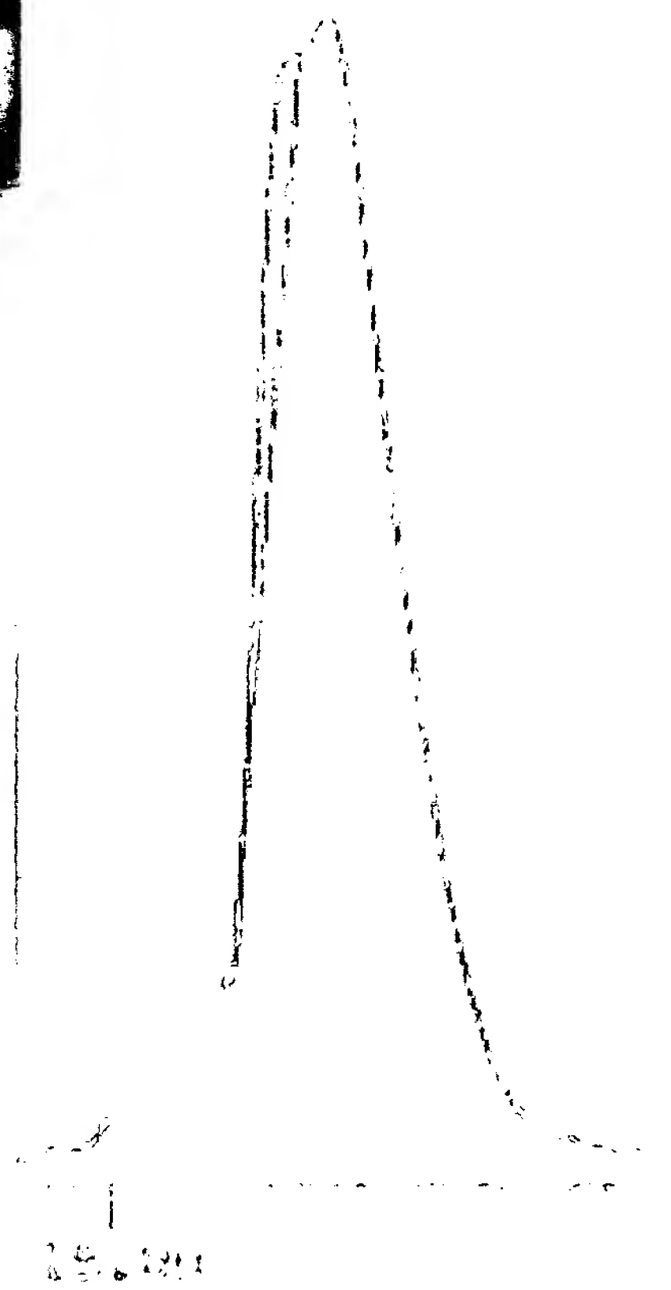


a

a. Mass spectra of C_{76}
b. HPLC traces (15.3 min.)
c. C_{76} solution in toluene

c



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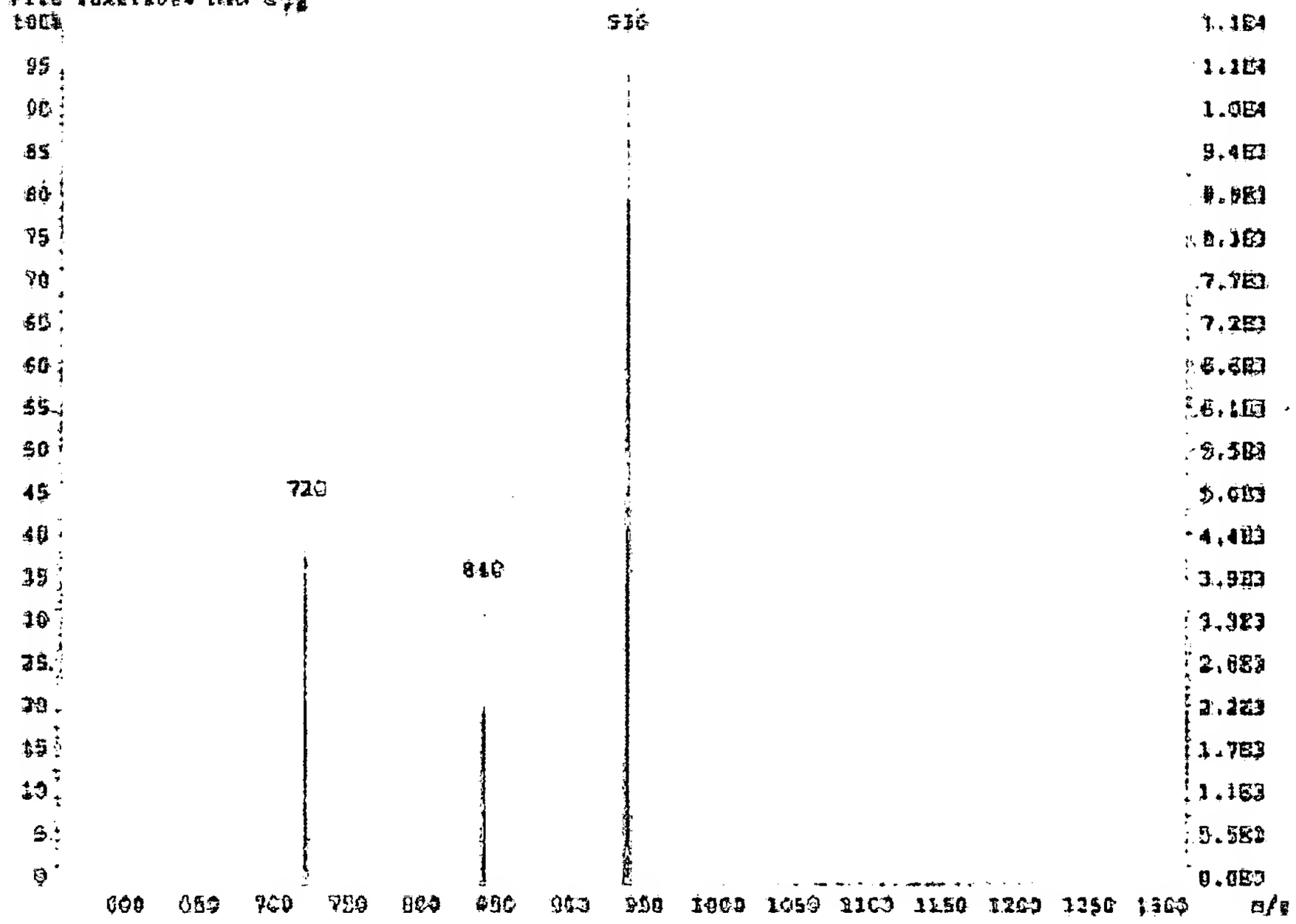
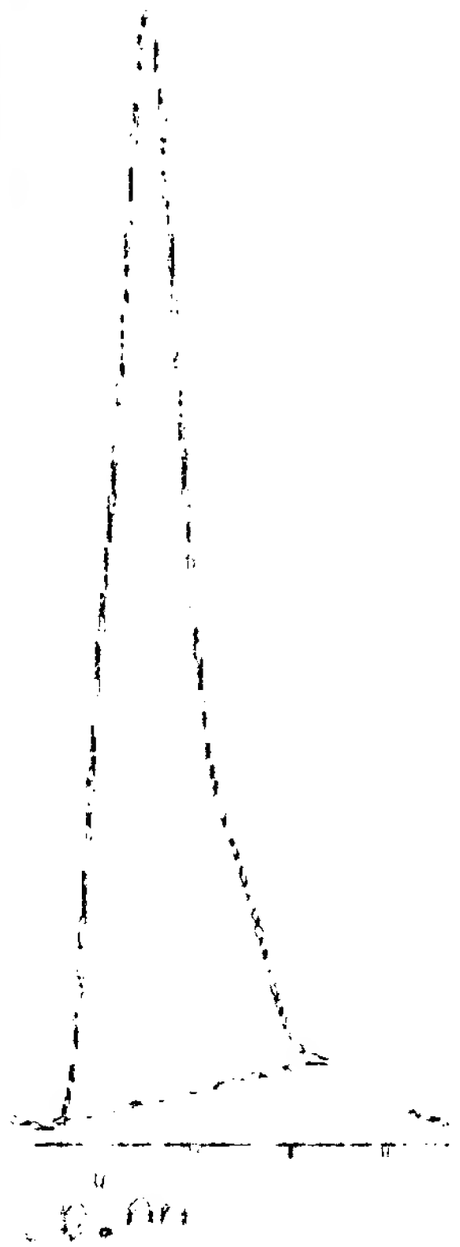


a

a. Mass spectra of C_{78}
b. HPLC traces (16.4 & 16.7.3 min.), two isomers
c. C_{78} solution in toluene.

c



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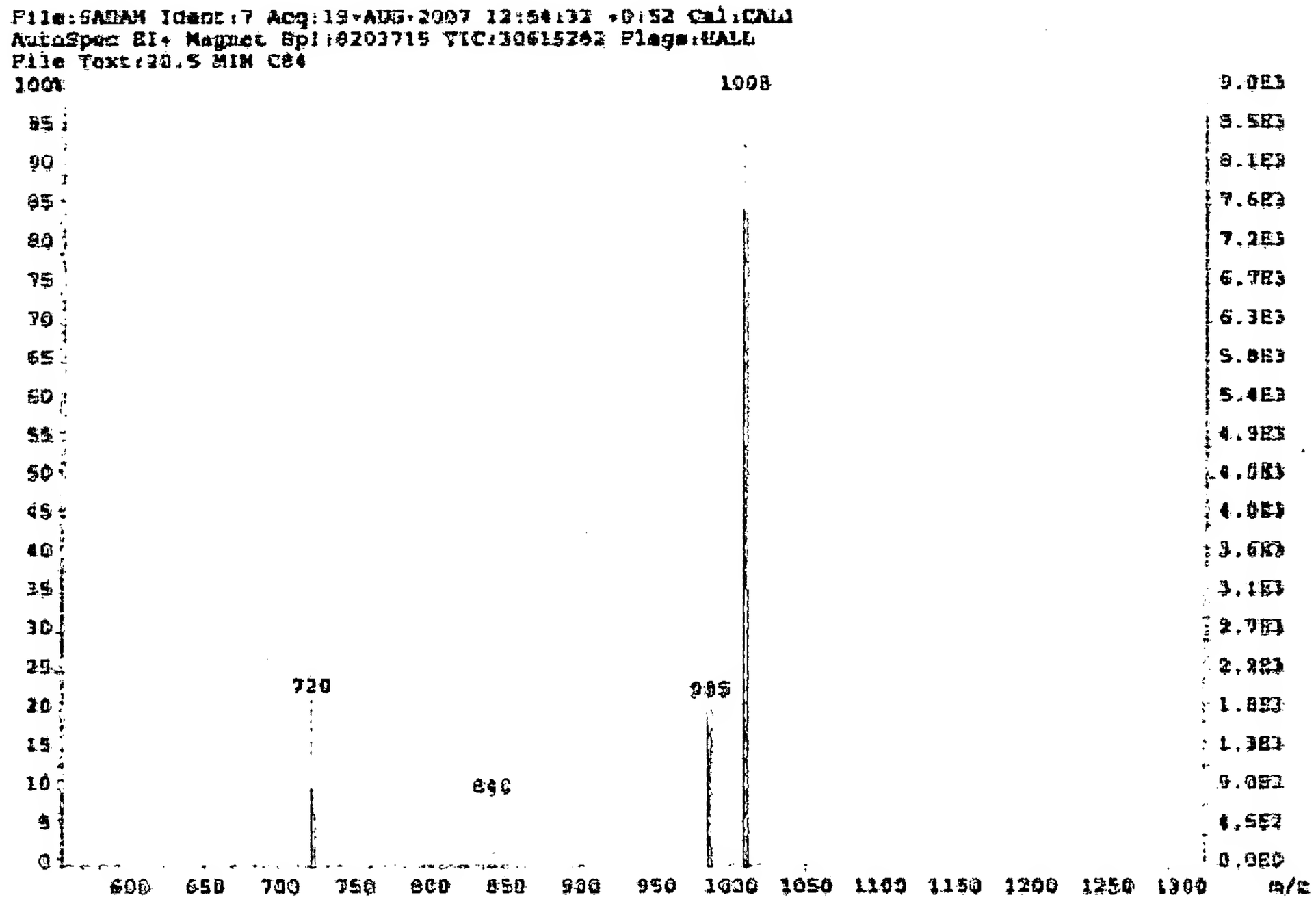
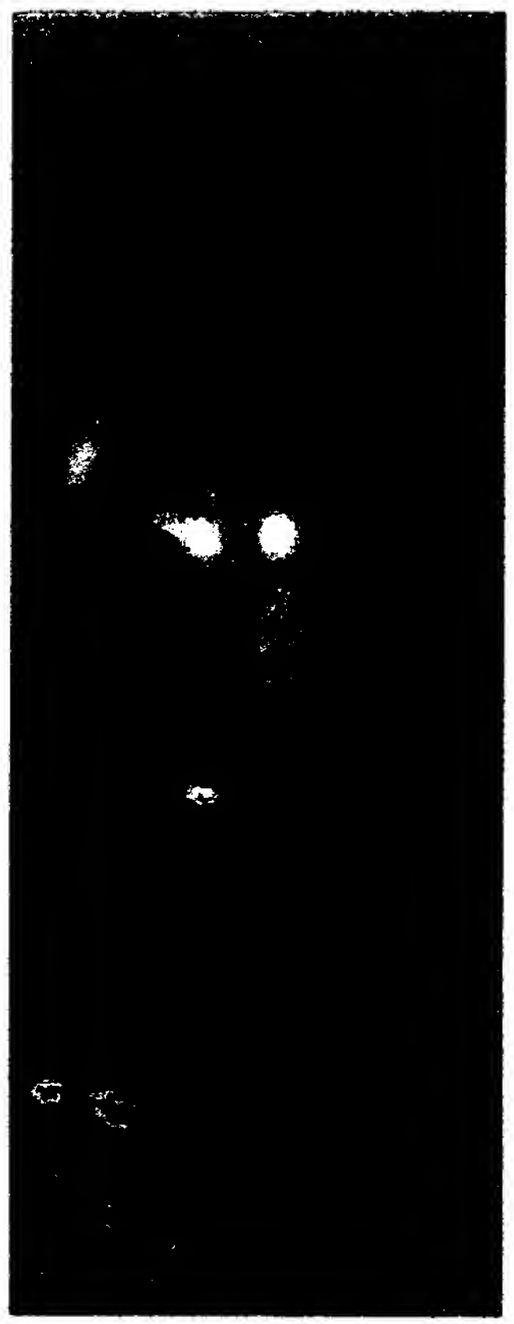
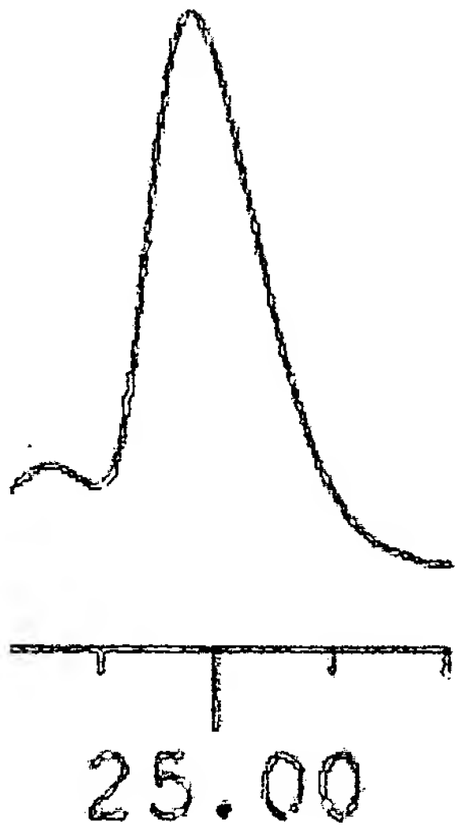
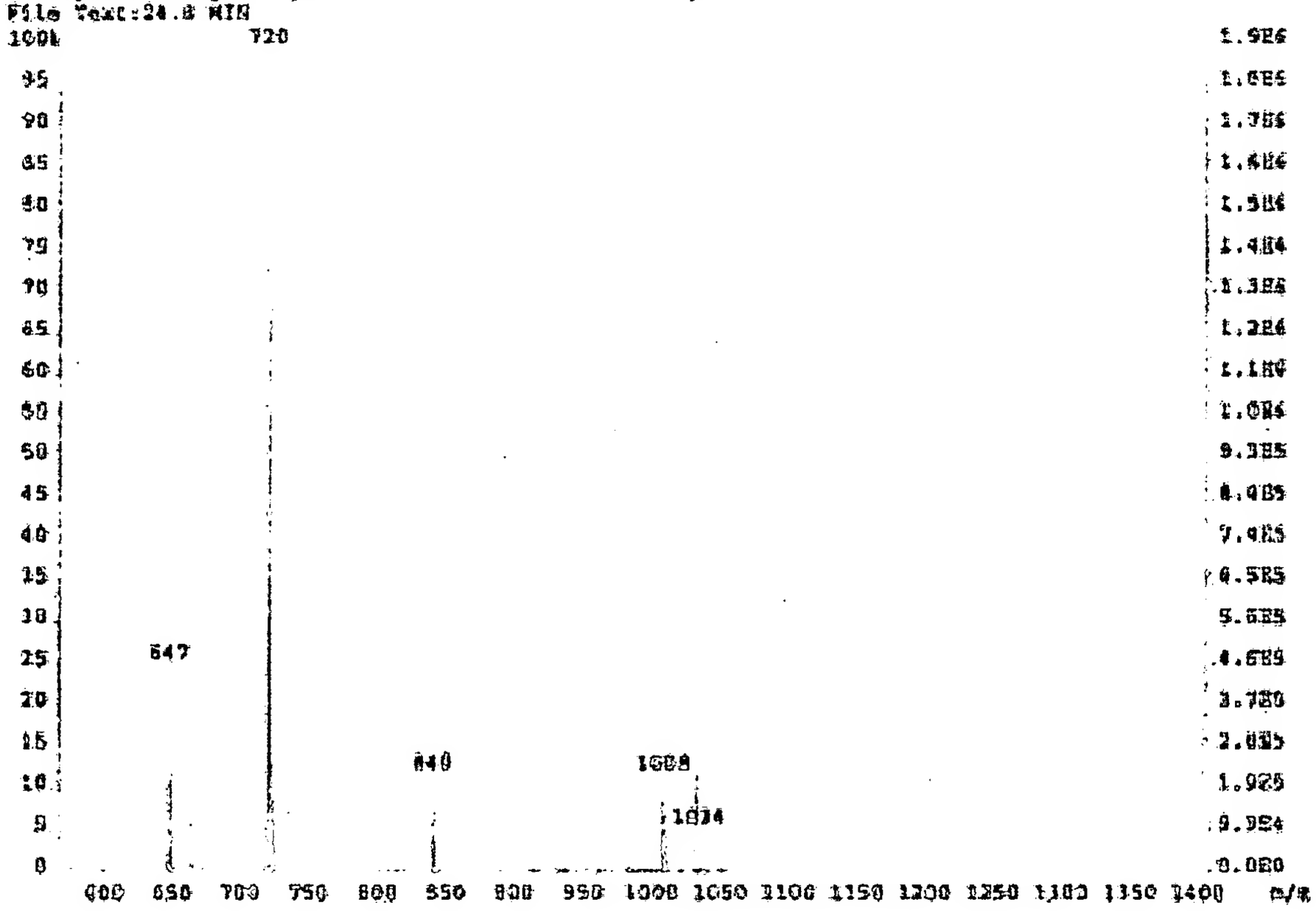


a

a. Mass spectra of C_{84}
b. HPLC traces (20.5 min.)
c. C_{84} solution in toluene







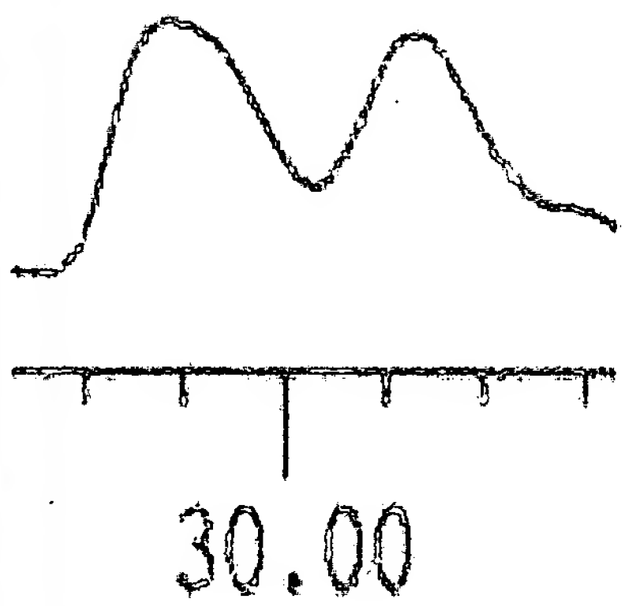
c

b

a

a. Mass spectra of C_{86}
b. HPLC traces (24.8 minutes)
c. C_{86} solution in toluene

c



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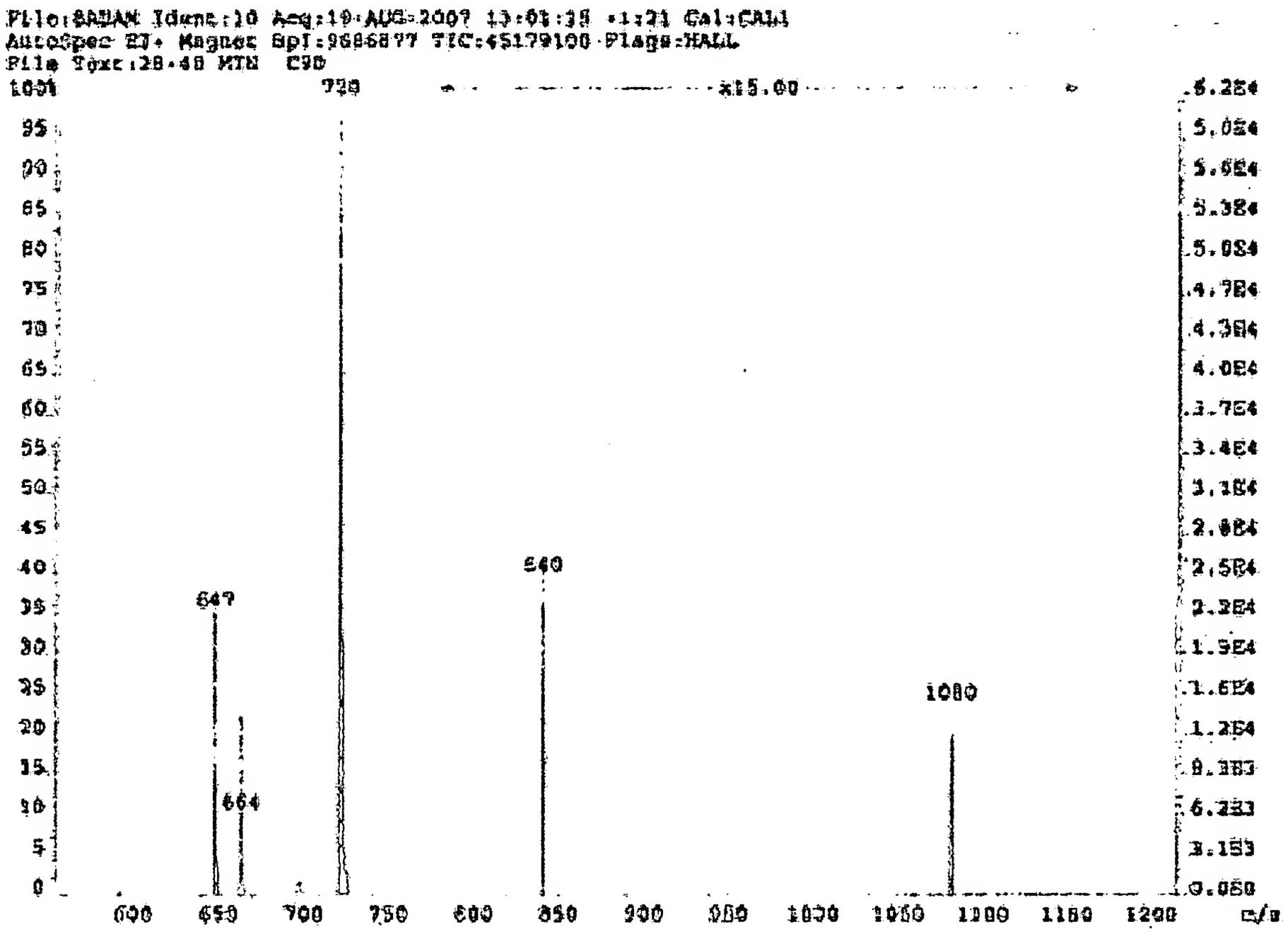
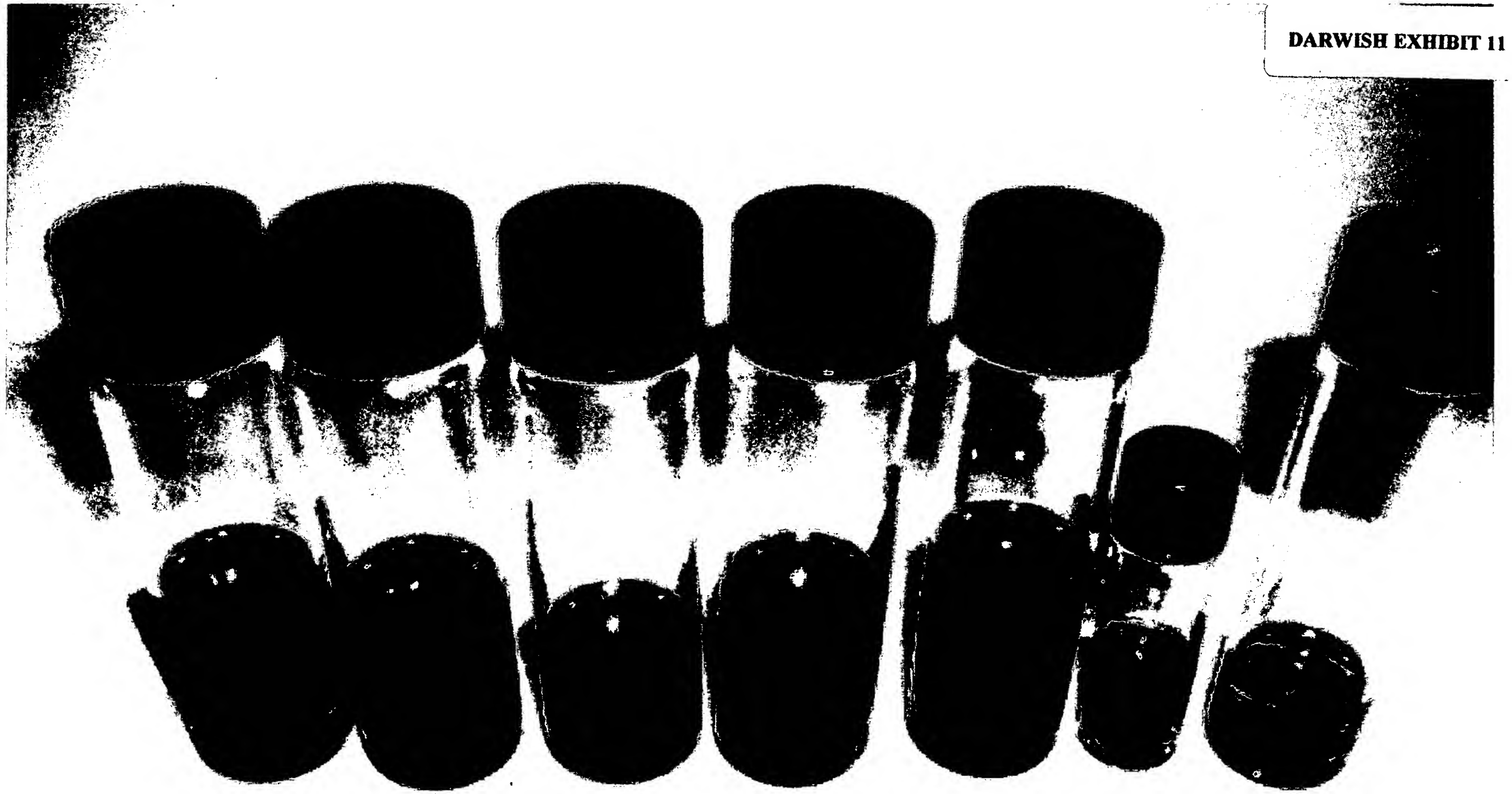


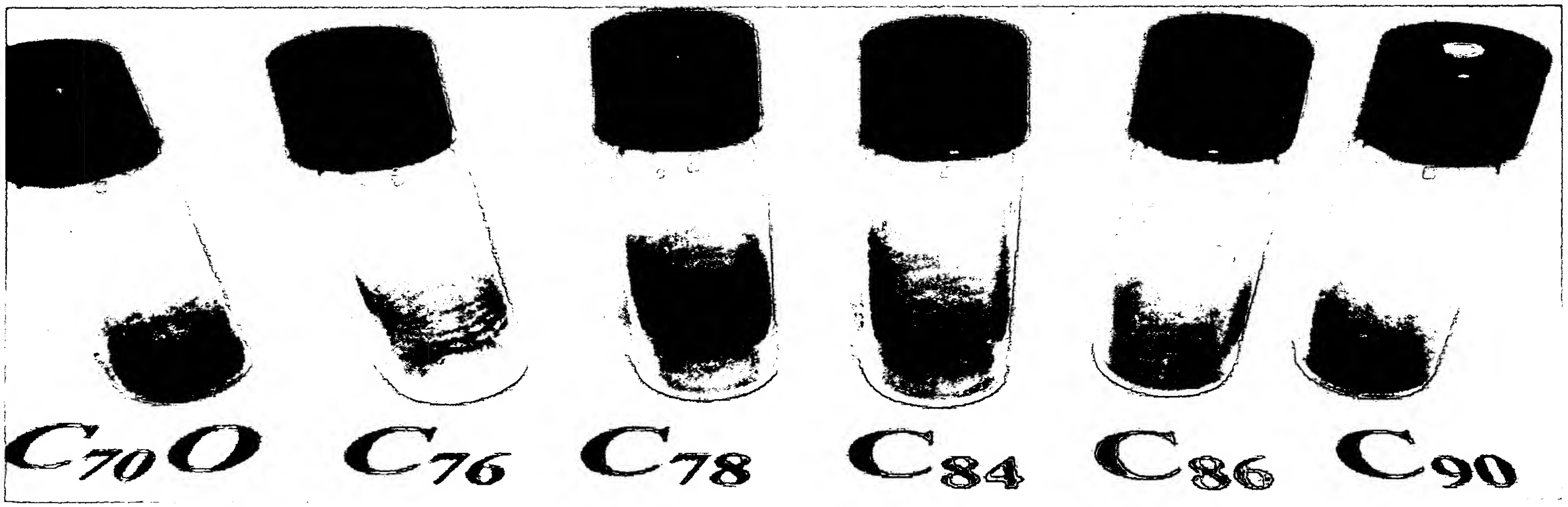
a

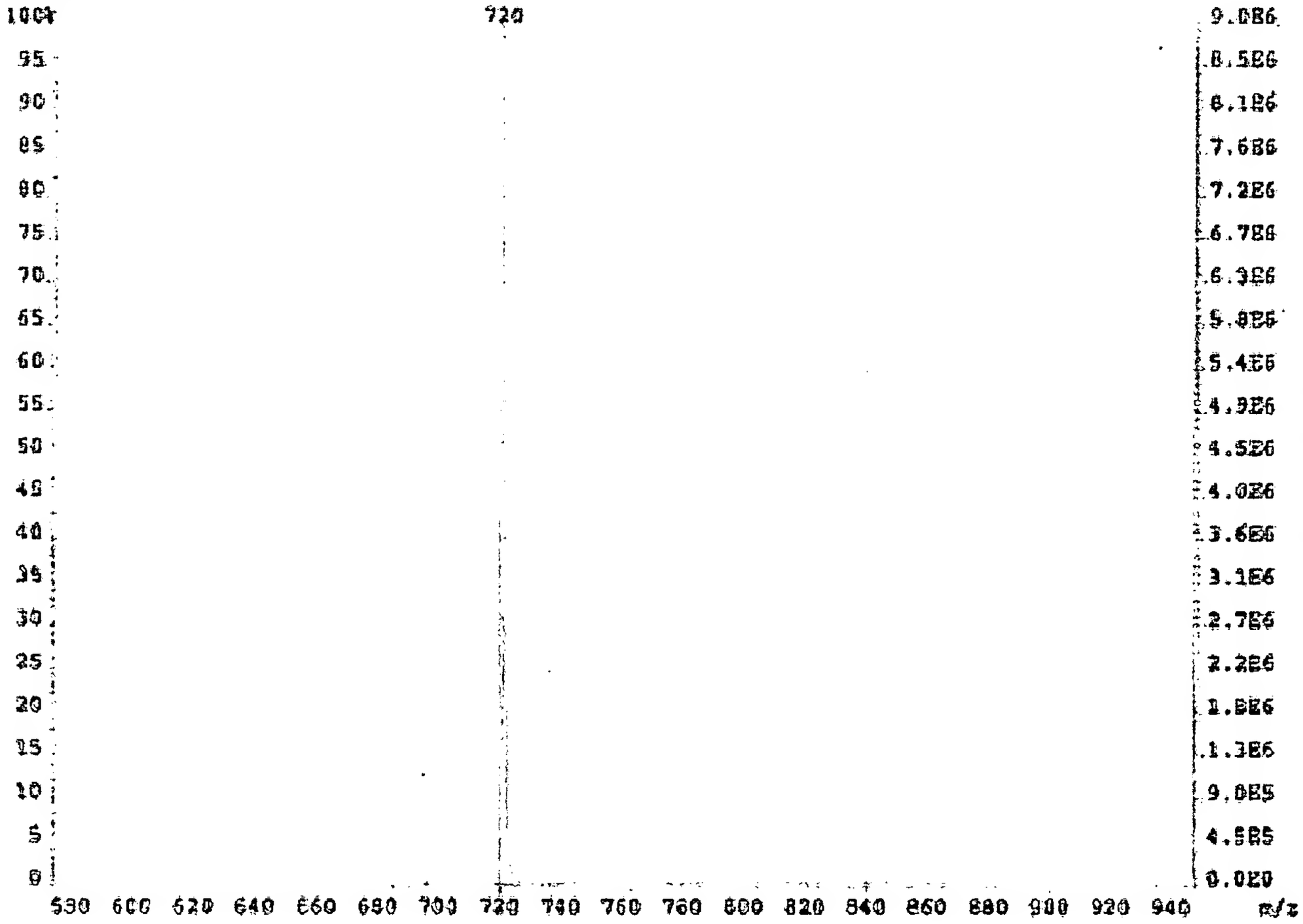
a. Mass spectra of C_{90}
b. HPLC traces (29 & 31minutes), two isomers
c. C_{90} solution in toluene

DARWISH EXHIBIT 11



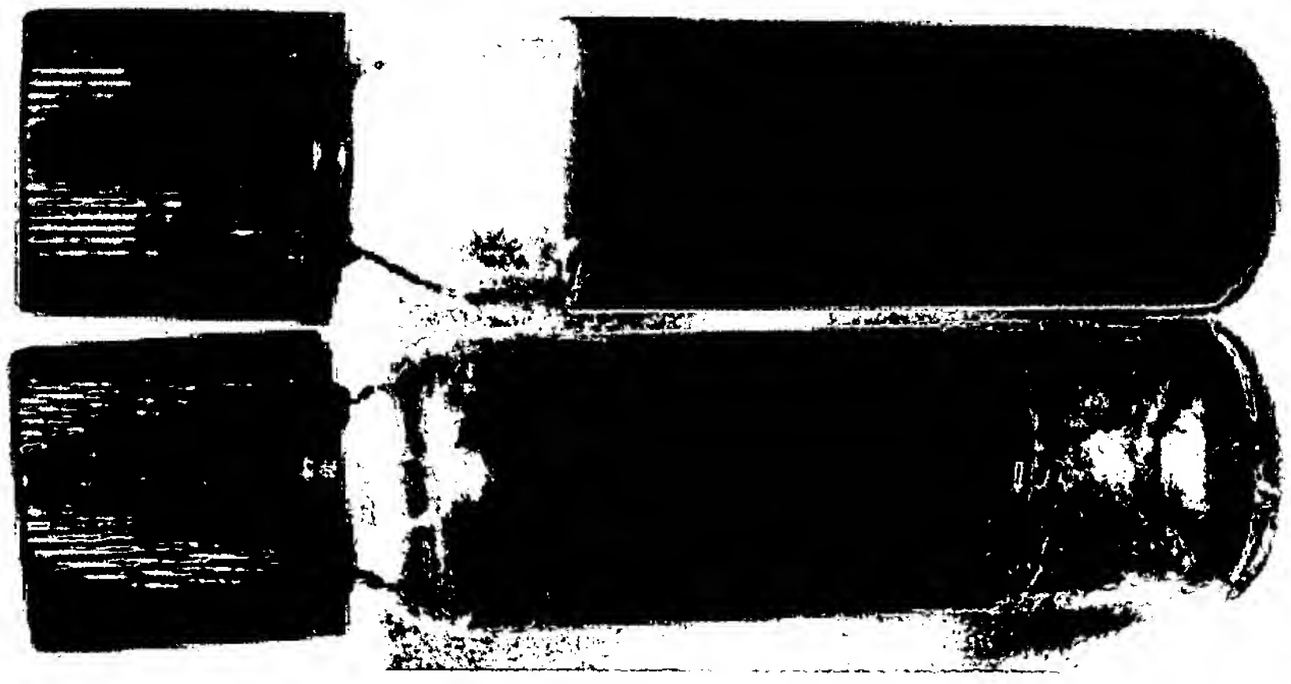
C_{60} C_{70} C_{76} C_{78} C_{84} C_{86} C_{90}

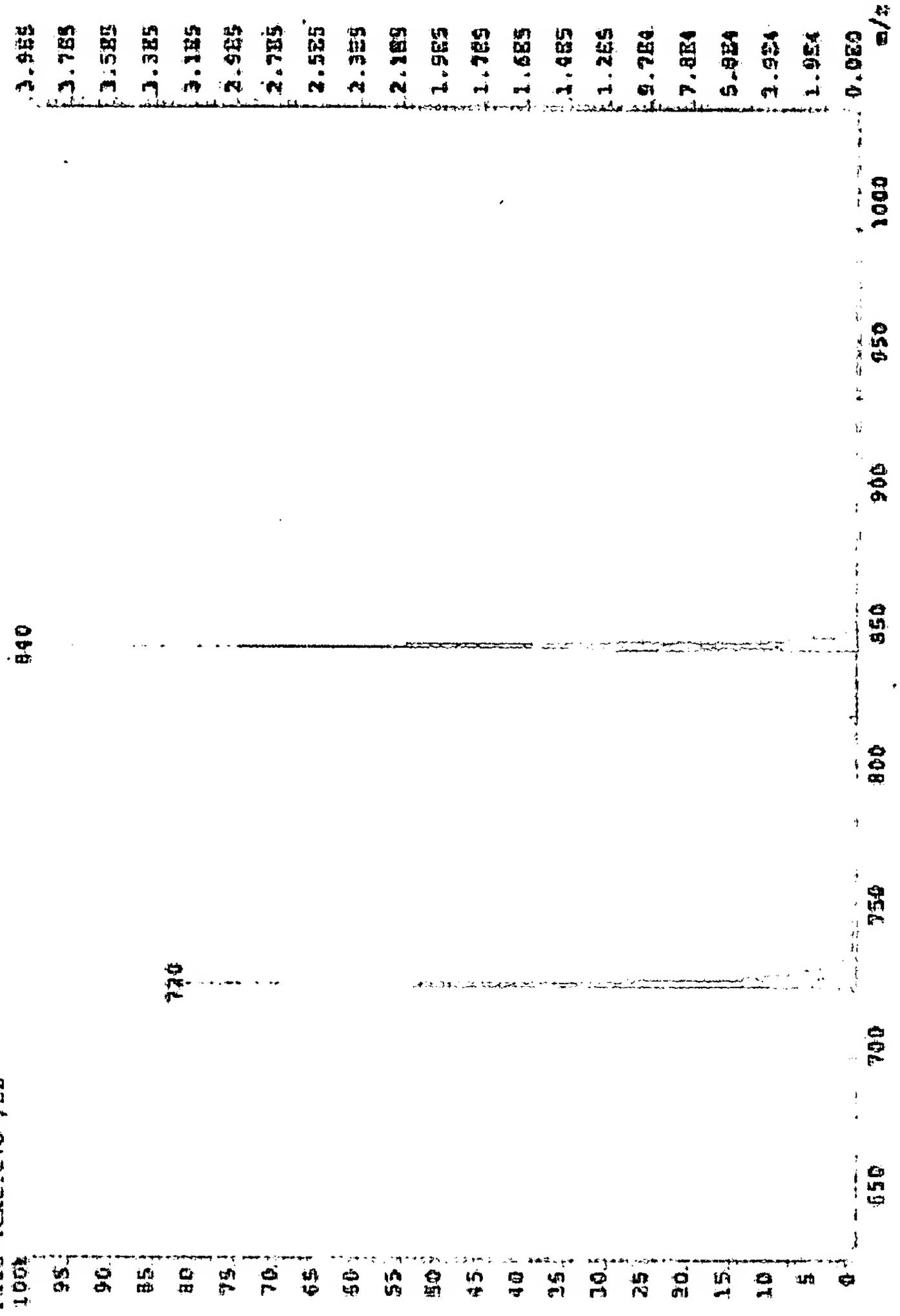




Mass spectra of C_{60}

C_{60} solution in toluene
C_{60} crystals

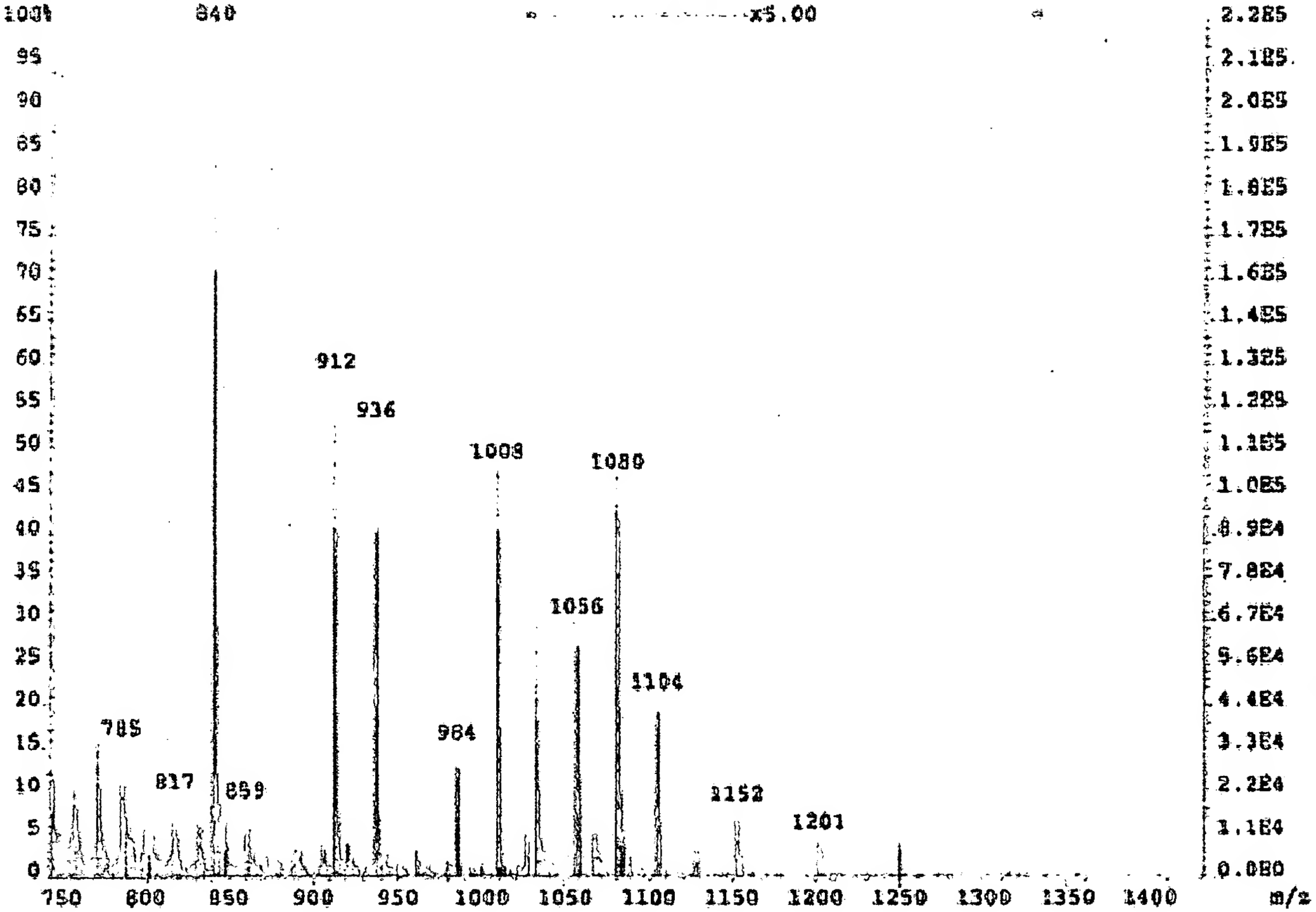




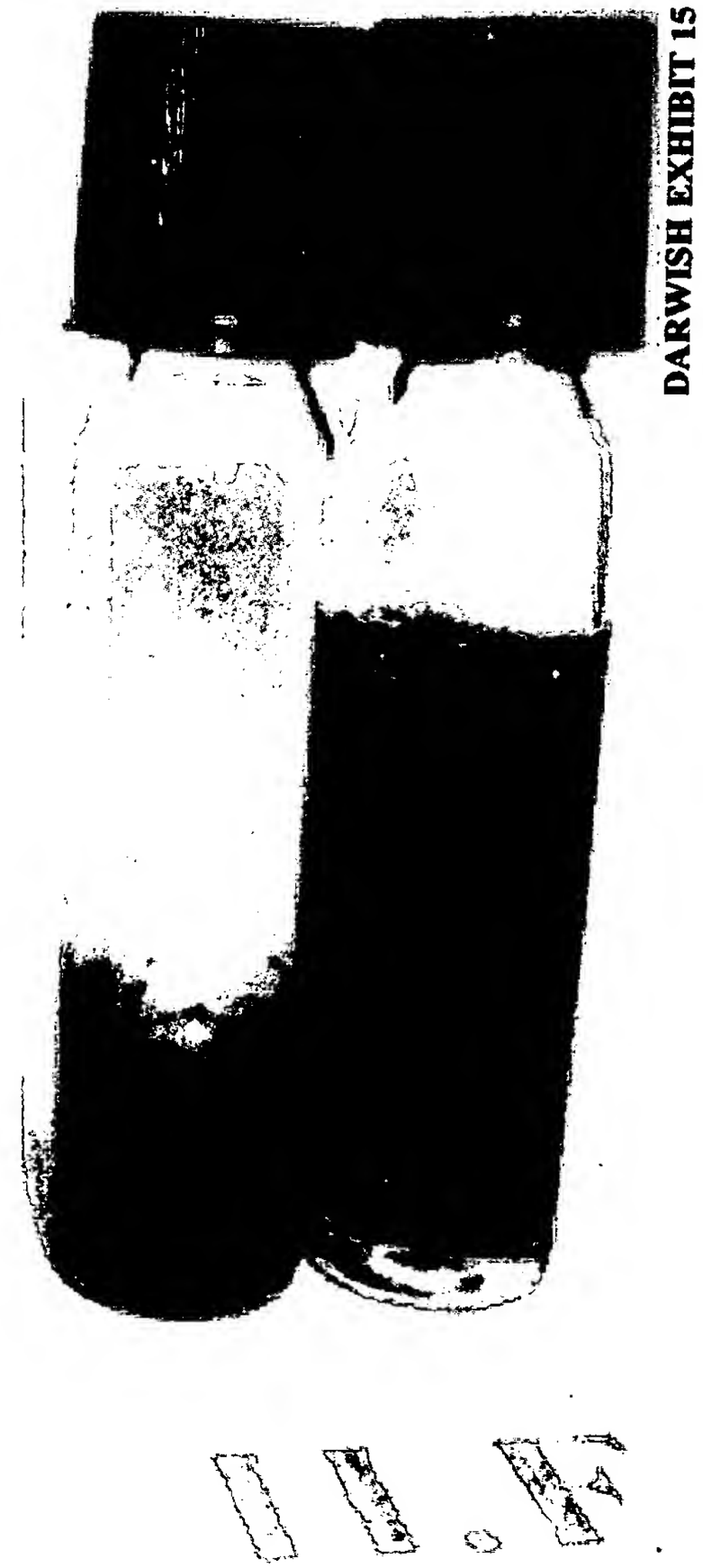
Mass spectra of C_{70}

C_{70} solution in toluene

C_{70} crystals

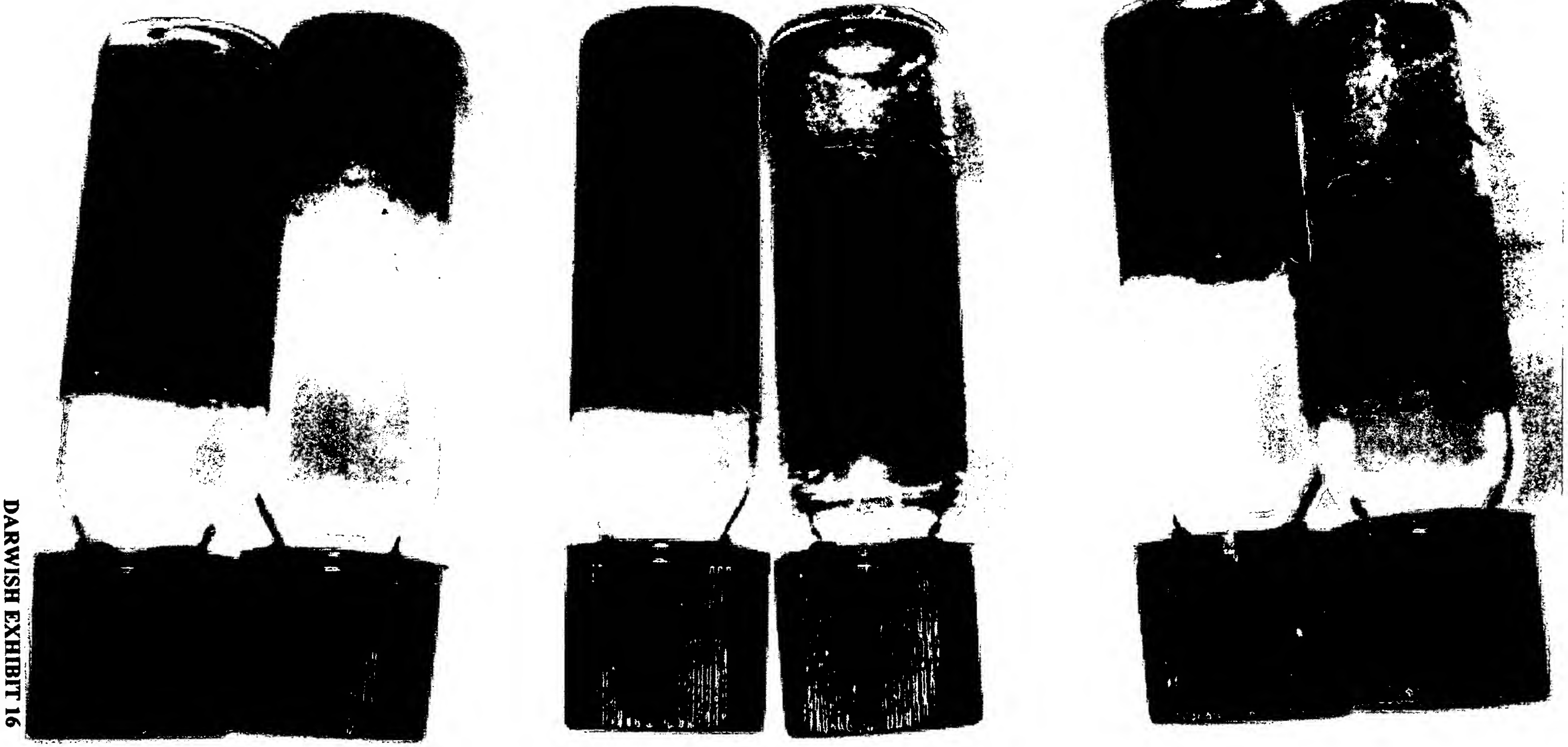


Mass spectra of H.F's up to C_{104}



H.F's solution in toluene

H.F's crystals



DARWISH EXHIBIT 16